UCRL-JC-121504 Abs

COMPARISON OF THE DISSOLUTION RATES OF URANIUM OXIDES IN AQUEOUS SOLUTIONS

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The purpose of our work has been to measure the intrinsic dissolution rates of uranium oxides under a variety of well-controlled conditions that are relevant to a geologic repository and allow for modeling. The intermediate oxide phase, U_3O_8, is quite stable and known to be present in oxidized spent fuel. Dehydrated schoepite, $UO_3 \cdot H_2O$, has been shown to exist in drip tests on spent fuel. U_3O_7 is stable in certain regimes.

Equivalent sets of U_3O_8 and $UO_3 \cdot H_2O$ dissolution experiments allowed us to examine systematically the effects of temperature (25-75˚C), pH (8-10) and carbonate (2-200x10^{-4} molar) concentrations at 8 ppm dissolved oxygen in the leaching solutions, equivalent to 0.2 atmosphere oxygen. Additional data on U_3O_7 at specified conditions were also obtained.

Results indicate that $UO_3 \cdot H_2O$ has a much higher dissolution rate than U_3O_8. Dissolution of $UO_3 \cdot H_2O$ shows a very high sensitivity to carbonate concentration. Present results show a 25 to 50-fold increase in room-temperature $UO_3 \cdot H_2O$ dissolution rates between the highest and lowest carbonate concentrations. This strong carbonate effect was demonstrated as well in earlier results on $UO_3 \cdot H_2O$ at low oxygen concentrations, which showed an even larger dissolution difference of almost 300 times. The intrinsic dissolution rate of unirradiated U_3O_7 and U_3O_8 is one to three times that of UO_2 under similar conditions.

Work performed under the auspices of the Yucca Mountain Site Characterization Project (YMP) and a USDOE/AECL (Canada) International Agreement as part of the Civilian Radioactive Waste Management Program by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

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